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HEALTH PHYSICS ASPECTS OF TRITIUM LUMINOUS DIAL PAINTING

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Abstract—The survey of a luminous dial painting plant is described. Under the conditions investigated, skin contamination was indicated as a major mode of entry of tritium into the body of the workers. A new unit, gram-Lambert (g-L) is proposed for comparison of radioactive luminous paints. It is shown that the benefit of the production of 1 g-μL is associated with the risk of approximately 0.16 mrem to the workers studied. A maximum tritium activity which may be processed by a worker without exceeding the maximum permissible body burden is proposed.

After severe injuries to people working with radium in the early days of luminous dial painting, numerous investigations were carried out to reduce the radiation dose to workers. In recent years efforts have been made to substitute other radionuclides for radium to reduce or eliminate external radiation by using radionuclides with no gamma radiation emission and reduce internal exposure by the application of radionuclides exhibiting a shorter biological half-life than radium. The radiation dose to the general public using wrist watches and other objects containing radioactive luminous paint may be substantially reduced if radium is replaced by other suitable radionuclides. In particular, the application of radium for production of high luminosity objects, such as safety devices for use in aircraft, could exhibit a prohibitively high ambient gamma radiation.

Among the substitutes proposed, only tritium and, ^{147}Pm to some extent, have found industrial applications.[1] The health physics considerations of tritium dial painting have been discussed by Vennart[1] and Jones and Lambert.[2] The latter investigators measured body burdens of workers in three different luminizing plants and concluded that the primary mode of entry of tritium into these individuals was by inhalation of tritiated water vapor. Vennart, in reviewing the results reported by Jones and Lambert, favored the use of radium over tritium and ^{147}Pm because of legal considerations and difficulties in collecting urine and fecal samples for radiological monitoring of the workers dealing with tritium and ^{147}Pm. Neither of these references deal in any detail with surface contamination.

This investigation is a part of the long-range evaluation of several aspects of luminous dial painting and deals with the health physics aspects of tritium dial painting. The present study was carried out in cooperation with the Georgia Department of Public Health in a plant located in the southeastern United States. The selected plant employs 15–20 workers and uses approximately 2000 Ci of tritium annually.

The luminous paint used consists of zinc sulfide coated with a tritiated polymer and mixed with an adhesive dissolved in an organic solvent. If thinning is desired, xylene is added to the mixture. The plant receives many grades of the active powder with specific activities of 9–250 mCi/g. If necessary, lower grades are prepared in a glove box by the addition of appropriate amounts of zinc sulfide. The preparation of the radioactive paint is also carried out in this glove box which is vented to the atmosphere outside the plant.

Several methods are used to apply the paint to

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the desired surfaces. These range from sophisticated rotary applicators to hand operated painting machines and manual painting. The painted objects are usually air dried but occasionally an oven is used to accelerate the drying process. All workers wear laboratory coats during working hours. No protective gloves are used in the plant. The workers are required to thoroughly wash their hands frequently during the day, particularly prior to coffee breaks, lunch, and leaving the plant. An ultraviolet light is used to monitor the workers' hands after handwashings to insure the removal of all visible traces of the paint. The plant is kept clean and is adequately ventilated. All hand painting is conducted under hoods in one room. Similarly silk screening, rotary machine operation, and drying of the wet paint are carried out in separate rooms. Monthly urine samples are collected from all workers and analyzed by a commercial radiobioassay laboratory.

SURVEY OF THE PLANT

Several surveys of the plant were carried out during this investigation. During the first visit the employees were informed of the objectives of the investigation. Also, the plant manager had previously discussed the importance of safety measures, such as regular urine analysis. Contrary to the experience reported by VENNART,[1] no problems were, nor had been, encountered in obtaining information or monthly urine samples.

The workers were interviewed and information relative to their food and water intake and the general condition of their health was obtained. All employees but one had a reasonably normal water intake; the one exception had 2–3 times the normal water intake in the form of soft drinks as a result of a prior kidney malfunction and the effects of the associated surgery.

A large number of smears was taken during survey. A 47-mm (0.8–2.5 μ pore size) microfilter was used which was soluble in toluene and exhibited little quenching in the subsequent liquid scintillation counting. In addition the solubility of the filter permitted the application of the internal standard with identical system properties (ISISP)[3] for counting efficiency determination.

Inasmuch as considerable air contamination of other plants had been reported[2], an air monitoring device was devised which distinguished between particulate, aqueous, and combustible (organic or elemental) forms of tritium. The general outline of the system is shown in Fig. 1. The air passes through a microfilter and drying column (DC I) where particulates (0.45 μ) and water, respectively, are separated. Tritium in elemental or organic form is combusted at 600°C in the presence of copper oxide and the resulting water is collected in a second drying column (DC II). The pump draws air through the system at a rate of 5–6 l./min. Finally a wet gas meter is used to integrate the air flow. Samples were collected at the normal breathing height of the workers.

Each room of the plant was surveyed by passing 100 l. of air through the system. The total air collection (including the replacement of drying columns) required about 30 min, making it possible to survey the entire plant in one day.

The microfilter and drying columns were then analyzed in the laboratory. Filter paper was analyzed in the same manner as smears. Drying columns were divided into three approximately equal parts; 150 ml of water was added to each and left for 24 hr to allow even distribution of the tritiated water. A portion of this water was subsequently distilled and analyzed by liquid scintillation counting. The entire system was evaluated in the laboratory prior to application in the plant. It was found that more than 90% of the added tritium as water vapor was collected in the first one-third of the columns. The same pattern was observed in the field.

Urine analysis was carried out by a method developed in this laboratory with a minimum limit of detection of 1–10 nCi/l. depending upon the condition of the urine.[4]

SURVEY RESULTS

Tritium concentrations in the air were generally below the maximum permissible concentrations (MPC) for a 40-hr week with body tissue as the organ of reference (5 nCi HTO/l.)[5] with values usually in the range of 2–3 nCi/l. Particulates constituted a maximum of 10% and usually only 5% of the total air activity.

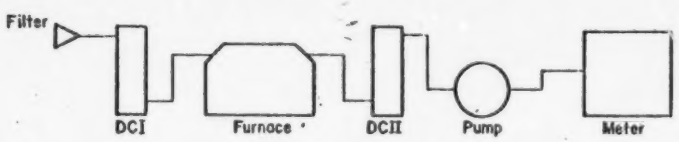


FIG. 1. Block diagram of the tritium air monitoring system. DC I and DC II are drying columns for the collection of tritiated water and the combustion products of elemental and organic tritium, respectively.

The major portion of tritium was collected as water. Considering the fact that tritium is incorporated in a polymer in the paint, it appears that tritium is released during the application of the paint, possibly through a hydrogen exchange with atmospheric moisture which seems to be in agreement with findings of JONES and LAMBERT.[2] Decomposition of the paint would have introduced tritium in comparable amounts in elemental or organic form which constituted in actuality only 5% of the total tritium in the air. The only exception was observed in the drying room in which a large amount of the solvent is evaporated and remains in the air, as the odor in that room clearly indicated. In this room the organic form of tritium was dominant. It is possible that in this case H-exchange occurs between organic solvent and the tritiated polymer. Although this exchange may be small in magnitude, the relative abundance of the solvent in the air results in 2–3 nCi/l. of combustible tritiated compounds.

Total air concentrations of 6–8 nCi/l. were observed in the small room in which the glove box is located and from which a large amount of paint is taken for distribution to the workers. Although these concentrations are slightly above the MPC for air, this room is used for distribution of the paint once or twice a day and is only occupied infrequently by two employees engaged in the distribution operation.

The tritium concentration in other working areas was relatively constant and fluctuated between 2 and 3 nCi/l. of air. This range remained relatively constant in several surveys made during this investigation.

A relatively high surface contamination was found in the plant, ranging from a few nCi to several μCi per wipe sample. All smears showed tritium contamination and represented door knobs, telephones, typewriters, and the surface of the soft drink machine in addition to working surfaces. A major difficulty in detecting and correcting such contamination lies in the fact that there is presently no feasible way for field measurement of tritium surface contamination of the order of a nCi/100 cm^2 or less. A regular smear analysis as carried out during this investigation is too expensive for routine application and, even more important, the time lag between collection of samples and transmission of results would not be compatible with the application of effective countermeasures.

The monthly urine analyses showed an apparent relationship between the total amount of tritium used in the plant and tritium body burdens of the employees. However, the body burdens were occasionally too high to be attributed to the air concentration of 2–3 nCi/l.

Of the two other modes of entry, namely, ingestion and diffusion through skin, the latter was suspected to make a major contribution to the body burden. All workers were required to wash their hands thoroughly prior to coffee breaks and lunch. Only 4 out of 15–20 workers were smokers and two of them admitted to smoking occasionally during their work. Also the application of cosmetics was done after coffee break and lunch. All this information was given by the employees to the surveyors during several surveys and was essentially confirmed by the manager and observed by the surveyors during the survey. Although there is no guarantee that these procedures were always followed, it is reasonable to assume that the contribution of ingestion to the body burden was smaller as compared to skin absorption, if one considers that all workers freely admitted not wearing gloves at any time. Another fact confirming this presumption is the comparison of body burden of two surveyors after the first survey. During this survey one surveyor (ET)

operated the air monitoring equipment while a second surveyor (AM) took smear samples and thus had a part of his skin surface in much closer proximity to tritium surface contamination. Neither of the surveyors used protective gloves. Subsequent urinalysis revealed a tritium concentration of 1.0 μCi/l. for AM and 5.0 nCi/l. for ET. Both surveyors wore laboratory coats, washed their hands prior to lunch and immediately after the completion of the survey. Neither one was a smoker. The possibility of ingestion of radioactivity was, in other words, small. Obviously the contribution of ingestion to the body burden of the workers cannot be separated from that of skin absorption. There is however an indication that skin absorption is the greater factor.

Subsequently, the plant underwent a general cleanup, including repainting. Also, plastic sheets were used to cover working surfaces and these sheets were replaced every week without changing other factors. Results of the next urinalysis demonstrated a dramatic change. Although the production of the plant, including tritium use, was not significantly changed, and the tritium concentration in the air remained essentially the same, body burdens dropped to 20–40% of the previous month, confirming the suspicion that tritium entry through the skin (and possibly ingestion) was a large contributing factor to the body burden.

Investigations at the Southeastern Radiological Health Laboratory[6] have shown that the biological half life (T_b) of the tritiated luminous paint introduced into the body of the rat is identical to the biological half life of tritiated water. If this observation is applied to man it is expected that after 30 days, or roughly three half lives ($T_b = 10$ days), approximately 10% of the activity is left. This must be taken into account to appreciate the reduction of new entries of tritium into the body of workers.

Animal investigations[7] have shown that the luminous paint diffuses through the skin and the maximum body burden is reached after several days and not after a few hours as in the case of water. This indicates the importance of cleaning the hands frequently and thoroughly.

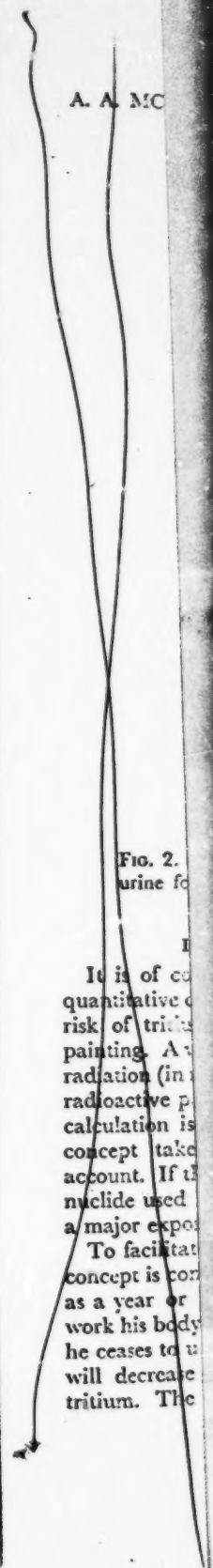
When the importance of skin contamination was recognized an intensive search was carried out to find usable gloves. The gloves tested were either inconvenient to wear for an 8-hr work period, or were not resistant to the solvents used. No commercially available, satisfactory gloves could be found.

As mentioned, an apparent correlation between the body burdens and the amount of tritium used was observed. In agreement with past experience,[2] the operators of rotary machines exhibited generally high body burdens although the tritium concentration in the air around the machines was always below the 5 nCi/l. limit.

The plant manager keeps records on the daily distribution of the paint to individual workers, making it possible to determine the tritium activity used by the workers for the period between any two urine sample collections. The correlation between these values and the urine concentrations as well as a least squares fit of the values for two time periods—namely, prior to and after cleaning of the plant—are shown in Fig. 2. Included in the same figure is the standard deviation of the values of the latter period.

Figure 2 may be used to estimate the maximum amount of activity which may be processed by individual workers without exceeding a certain body burden.

There is presently no internationally accepted limit for the tritium concentration in the body.[1] The National Committee for Radiation Protection proposed a tritium concentration of 30 μCi/l. of water in body tissue for a continuous exposure. This value would result in an annual dose of 5 rem[8] if an even distribution of tritium in the total body is assumed. In the United States, values between 28–30 μCi/l. are used as maximum permissible concentration of tritium in body water. Figure 2 shows that for the latter period of the survey a body burden of 30 μCi/l. of body water corresponds to an individual monthly usage of approximately 20 Ci of tritium. This value may be used as a guideline for the surveyed plant. The availability and application of protective gloves could increase the usage level considerably without a corresponding increase in body burden. It is probable that similar values may be obtained for other plants, although under different conditions this relationship may vary.



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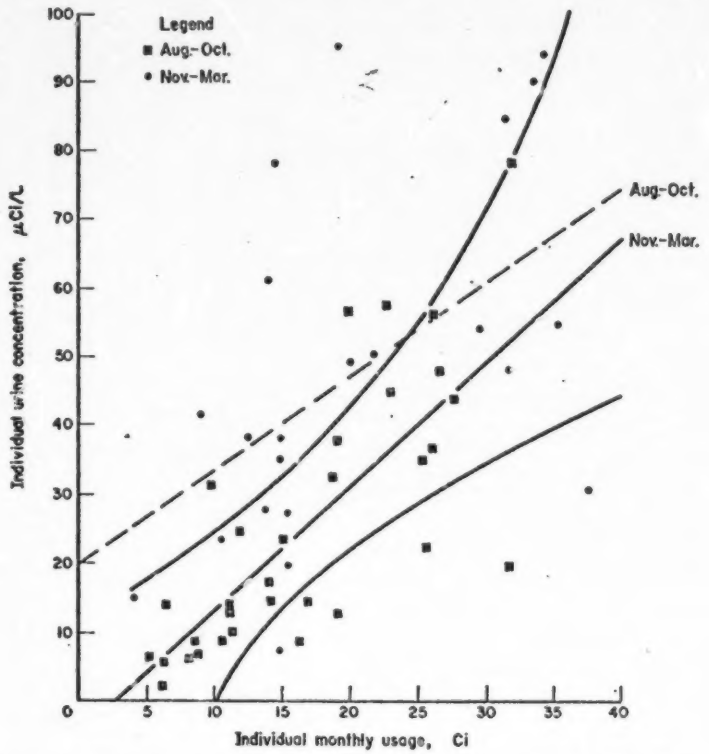

FIG. 2. Correlation between the individual monthly usage and the tritium concentrations in urine for the periods between August–October and November–March as well as the standard deviation (solid curves) of the latter values.

DOSE CALCULATION

It is of considerable interest to establish a quantitative correlation between the benefit and risk of tritium application in luminous dial painting. A worker receives a certain amount of radiation (in rem) as a result of working with the radioactive paint. The proper concept for dose calculation is that of dose commitment. This concept takes the post exposure dose into account. If the biological half life of the radionuclide used is long, the latter may constitute a major exposure to the worker.

To facilitate the calculation, the risk–benefit concept is considered for a prolonged time such as a year or more. When a worker starts to work his body burden rises gradually and when he ceases to use tritiated paint his body burden will decrease with the biological half life of tritium. The body burden in the intermediate time will fluctuate depending upon working conditions and possible accidental intake. For the purpose of dose calculation, the average of the body burden of all workers during the entire survey period is used. The averaging process disregards the individual variations and is used in this paper solely to evaluate the overall risk of the operation.

The application of the concept of dose commitment requires that the dose received by the worker after he ceases to work be considered. In the case of tritium it is assumed that this additional dose is negligible as compared to the annual dose during his employment. This assumption, although an approximation, is justified by the short biological half life of tritium. The exposure after termination of work contributes only a small dose as compared to the annual dose during the working period. In

addition, this underestimation of the dose is partially accounted for by the overestimation of the dose during the build-up period.

The records of the plant manager indicate that within the period covered by this investigation an average of 146 Ci of tritium per month was applied by an average of 16.8 employees. The results of the analyses reveal an average tritium concentration in urine for the entire period of 20.4 μCi/l. If these values are extended to a year it can be calculated that every worker would process

$$\frac{146(\text{Ci/month}) \times 12(\text{months})}{16.8\ (\text{workers})} = 104.3$$

Ci tritium and would receive a dose of

$$\frac{20.4(\mu\text{Ci/l.}) \times 5(\text{rem})}{30(\mu\text{Ci/l.})} = 3.4\ \text{rem.}$$

WALLHAUSEN[9] reports that 1.3 Ci of tritium used in one gram of luminous paint produce a light equivalent of 100 microlamberts (μL). According to the information of the manufacturer of the luminous paint, improvements in the production of the paint in recent years have decreased this value to 0.2 Ci/g for a luminosity of 40 μL which is the highest grade used in the surveyed plant. Accordingly, in the following calculations the manufacturer's value is used.

In order that the above values may be used for the risk-benefit calculations, the amount of radioactivity processed by the workers must be converted to a unit representing the total light emitted by the produced luminous paint. Lambert (L) is the unit of light density in candela (cd)/πcm^2 and, as such, is proportional to the specific activity of the paint and thus unsuitable for such a consideration. It is desirable to determine the light intensity of the radioactive paint in cd or a unit reasonably well related to it. The product gram-Lambert (g-L) seems to be such a unit. This unit is defined as the light produced by 1 g of paint distributed over 1 cm^2. Inasmuch as the specific weight of zinc sulfide is approximately 4, the thickness of the paint will be 2.5 mm. This definition of g-L is useful in comparing radionuclides for their suitability for luminous dial painting. Based on above mentioned values, the benefit of the production of one gram-microlambert is associated with the risk of

$$\frac{3.4 \times 10^3(\text{mrem}) \times 0.2(\text{Ci})}{104.3(\text{Ci}) \times 40(\text{g-}\mu\text{L})} = 0.16\ \text{mrem.}$$

If the period prior to the cleanup of the plant is used 1 g-μL/corresponds to 0.18 mrem, whereas the value for the period subsequent to the cleanup is approximately 0.13 mrem.

The risk-benefit consideration expressed in mrem/g-μL as discussed in this paper for tritium requires the knowledge of the radiation dose (in mrem) received by the workers as a result of processing a certain amount of radioactive luminous paint. This value is, however, presently not available for the two other radionuclides commonly used, i.e. radium and promethium. The availability of these values would permit a comparison for health physics purposes among these radionuclides for application in luminous dial painting.

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Mention of commercial products used in connection with the work reported in this article does not constitute an endorsement by the Public Health Service.

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